Coherent Control for a Two-level System Coupled to Phonons

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The interband polarizations induced by two phase-locked pulses in a semiconductor show strong interference effects depending on the time τ_1 separating the pulses. The four-wave mixing signal diffracted from a third pulse delayed by τ is coherently controlled by tuning τ_1. The four-wave mixing response is evaluated exactly for a two-level system coupled to a single LO phonon. In the weak coupling regime it shows oscillations with the phonon frequency which turn into sharp peaks at multiples of the phonon period for a larger coupling strength. Destructive interferences between the two phase-locked pulses produce a splitting of the phonon peaks into a doublet. For fixed τ but varying τ_1 the signal shows rapid oscillations at the interband-transition frequency, whose amplitude exhibits bursts at multiples of the phonon period.

Ultrafast spectroscopy on semiconductors gives a direct insight into the loss of coherence between optically excited carriers. The dephasing takes place through different mechanisms such as emission of phonons, Coulomb interaction or scattering by impurities. In the first hundred femtoseconds after an optical excitation few scattering processes have occurred and the electronic wavefunction may be described by a superposition of different states with well defined phase relations. The resulting quantum beats are a signature of phase coherence.

Quantum beats with the longitudinal optical (LO) phonon frequency have been observed in four-wave mixing (FWM) experiments on GaAs which was excited resonantly at the excitonic energy[1]. The beatings are due to interference between states with different number of virtual phonons. Indeed electrons close to the band edge which do not have enough energy to emit any real LO phonon, form a polaronic state.

The phase coherence of the excitation may also be used to coherently control the optical response of the sample via a pair of phase-locked pulses which induce interfering polarizations in the sample[2]. Recently this coherent control was applied to the quantum beats in the FWM signal[3]. Depending on the time delay τ_1 between phase-locked pulses the amplitude of the quantum beats was modulated, thus experimentally demonstrating that the phase coherence is not lost in the first few scattering processes.

The present work computes the FWM response in third order in the field for a simple two-level system treating exactly the coupling to a single phonon mode. The calculations closely follow Ref. 4 which studied the linear response in connection to phonon broadening of impurity spectra in semiconductors[5]. Recently the FWM response in the same model was obtained for δ-function pulses by solving the infinite hierarchy of kinetic equations[6].

We focus here on the regime of strong coupling between electrons and phonons and discuss the recent coherent-control experiment on the polar material ZnSe, where peaks separated by approximately half the phonon period were observed and interpreted as signatures of two-phonon processes[7]. In the two-level system, however, we attribute these features to a splitting of the phonon peaks into doublets due to destructive interference between the signals generated by the two phase-locked pulses. We further predict non-perturbative bursts of the FWM signal as a function of delay between phase-locked pulses at multiples of the phonon period T_{ph}.

A two-level system $\mid i \rangle$, $i = 0, 1$ with energies $\epsilon_0 < \epsilon_1$ couples to the position $a + a^\dagger$ of a harmonic oscillator of frequency Ω with a level-dependent coupling strength g_i. In addition the classical light field $E(t)$ induces transitions between the two levels:

$$H = \sum_{i=0,1} \left(\epsilon_i + g_i(a + a^\dagger) \right) \mid i \rangle\langle i \mid + \Omega a^\dagger a$$
$$- \mu E(t) \left(\mid 0 \rangle\langle 1 \mid + H.c. \right). \qquad (1)$$

This model describes the coupling of electrons to LO phonons of frequency $\Omega = 2\pi/T_{ph}$ using a single oscillator. The two levels may be viewed as states in the valence and conduction bands which are resonantly excited by light. Within this interpretation the coupling constants g_0 and g_1 should be taken equal since the Fröhlich interaction does not depend on the effective mass of the electrons i.e. on the band[8]. Another picture associates the level $\mid 0 \rangle$ to the full valence and empty conduction band while $\mid 1 \rangle$ corresponds to an excitonic state which has an effective coupling[9] g_1 to the phonons differing from the ground-state value g_0. This interpretation is particularly meaningful for the FWM experiments where the excitation is resonant with the excitonic level.

The model considers only scattering of the electron within the same level and hence cannot describe relaxation effects. This approximation which neglects the re-

coil of the electron is particularly relevant to the strong coupling regime since it corresponds to the small-polaron limit[4,10].

Four-wave mixing. A first pulse $E_1(t)$ creates an interband polarization in the sample while a second pulse $E_2(t-\tau)$ delayed by a time τ is diffracted along the direction $2q_2 - q_1$ where q_1 and q_2 are the propagation directions for the first and second pulses, respectively (see inset of Fig.1). The FWM signal $F(t,\tau)$ at time t and delay τ is related to the third order response[11] $\chi^{(3)}$:

$$F(t,\tau) = -i\mu^3 \int_{-\infty}^{\infty} dt_1 dt_2 dt_3 E_1^*(t_2)$$

$$E_2(t_1 - \tau)E_2(t_3 - \tau)\chi^{(3)}(t - t_1, t - t_2, t - t_3) \quad (2)$$

The electron occupies the lowest level $\mid 0\rangle$ before the pulses arrive, and the oscillator is in thermal equilibrium at an inverse temperature $\beta = 1/k_B T$. The pulses cause transitions between electronic levels which affect the evolution of the oscillator via the level dependent Hamiltonian $H_i = \Omega a^\dagger a + \epsilon_i + g_i(a + a^\dagger)$. The response function involves a thermal average of evolution operators and has a main contribution at positive time delays and a second term for $t_1 > t_2 > t_3$ i.e. when the two pulses overlap:

$$\chi^{(3)}(t_1, t_2, t_3) = \Theta(t_1)\Theta(t_2 - t_1) \times$$

$$\left(\Theta(t_3)\langle e^{iH_1(t_2-t_1)}e^{iH_0 t_1}e^{-iH_1 t_3}e^{iH_0(t_3-t_2)}\rangle + \right.$$

$$\left. \Theta(t_3 - t_2)\langle e^{-iH_1 t_1}e^{iH_0(t_2-t_1)}e^{iH_1(t_2-t_3)}e^{iH_0 t_3}\rangle\right). \quad (3)$$

The evaluation of the terms in brackets follows exactly Ref. 4. The Hamiltonian H_1 is diagonalized by the Lang-Firsov transformation[12] $S = g(a^\dagger - a)$: $\exp(S)H_1\exp(-S) = H_0 + \epsilon$ where $\epsilon = \epsilon_1 - \epsilon_0 + (g_0^2 - g_1^2)/\Omega$ is the renormalized transition energy and $g = (g_1 - g_0)/\Omega$ a dimensionless coupling constant. Since only the difference $g_1 - g_0$ enters the transformation, it is essential that e.g. excitonic effects renormalize the coupling strengths from the non-interacting value $g_1 = g_0$.

The product of evolution operators in Eq.(3) is evaluated using the previous canonical transformation and the following relation between the time-dependent operators $S(t) = \exp(iH_0 t)S \exp(-iH_0 t)$:

$$e^{S(t_1)}e^{-S(t_2)} = e^{S(t_1)-S(t_2)}e^{-ig^2\sin(\Omega(t_1-t_2))}. \quad (4)$$

We do not reproduce here the lengthy expression for the response function but instead discuss the FWM signal $F(t,\tau)$ for δ-function pulses. The signal oscillates at the renormalized energy ϵ with a time delay 2τ typical of a photon echo. The polaronic nature of the state shows up in the exponential dependence on the coupling constant, and the temperature enters via the Bose function $N_B = 1/(\exp(\beta\Omega) - 1)$:

$$F(t,\tau) = -i\Theta(\tau)\Theta(t-\tau)e^{-i\epsilon(t-2\tau)} \times$$

$$e^{-g^2\left((N_B+1/2)|2e^{i\Omega\tau}-e^{i\Omega t}-1|^2 - 2i\sin(\Omega\tau)+i\sin(\Omega t)\right)}. \quad (5)$$

The FWM signal which depends simply on $\Omega\tau$ exhibits quantum beats at harmonics of the bare LO frequency, in contrast to the frequency shift seen experimentally[1]. While the intraband scattering is missing in the present exactly solvable model, the simulations of Ref. 1 for a full two-band model do reproduce the correct frequency within approximate quantum kinetic equations. Moreover the FWM signal in Eq.(5) has evenly spaced phonon replica in the frequency domain, which are not observed in experiments.

Coherent control. Two phase-locked pulses of equal amplitude propagate in direction q_1 and are separated by a time delay[13] τ_1 as shown in the inset of Fig. 1. Since the FWM is linear in the field $E_1(t)$, the total signal is simply $F(t,\tau) + F(t + \tau_1, \tau + \tau_1)$ which shows strong interference effects depending on τ_1 through the relative phase of the induced polarizations.

The time-integrated FWM $I(\tau, \tau_1) = \int_{-\infty}^{\infty} \mid F(t,\tau) + F(t + \tau_1, \tau + \tau_1) \mid^2 dt$ can be calculated analytically for δ-function pulses, $E_1(t) = \delta(t) + \delta(t + \tau_1)$. A phenomenological lifetime Γ which accounts for the damping of the polarization is introduced as an imaginary part of ϵ. For simplicity we give here only the result at zero temperature ($N_B = 0$) and for $\Gamma \ll \Omega$, which involves the modified Bessel function I_0:

$$I(\tau, \tau_1) = G(\tau, 0) + G(\tau + \tau_1, 0) + 2G(\tau + \frac{\tau_1}{2}, \tau_1). \quad (6)$$

$$G(\tau, \tau_1) = \frac{e^{-2\Gamma\tau}}{2\Gamma}Re\left[e^{-i\epsilon\tau_1 - 2g^2(3 - 2e^{-i\Omega\tau_1/2}\cos(\Omega\tau))} \times \right.$$

$$\left. I_0\left(2g^2\left((2 - e^{i\Omega(\tau-\tau_1/2)})(2 - e^{-i\Omega(\tau+\tau_1/2)})\right)^{1/2}\right)\right]. \quad (7)$$

We first discuss the contribution of a single pulse when the integrated FWM equals $G(\tau, 0)$ which decays exponentially with time delay and is modulated by terms oscillating with multiples of Ω. In the weak coupling regime, only the first harmonics $\cos(\Omega\tau)$ contributes to the modulation as given explicitly in Ref. 6. Figure 1 shows the integrated FWM response $I(\tau, \tau_1 = 0)$ calculated with the exact $\chi^{(3)}$ for pulses of finite width which are taken as sech^2-shaped pulses resonant with the two levels and of 13fs duration. The parameters[7] for ZnSe are $\Omega = 32$meV, $\epsilon = 2.8$eV, $T = 77$K and $1/\Gamma = 300$fs. The weak modulation of the signal for a small coupling $g = 0.2$ relevant to GaAs, evolves into distinct peaks at multiples of the phonon period for a larger coupling $g = 0.8$ as in ZnSe. In the strong coupling regime indeed, the many harmonics sum up into sharp peaks of width proportional to $1/\sqrt{g}$: $G(\tau, 0) \simeq \exp(-\Gamma\tau - 16g^2\sin^4(\Omega\tau/2))$.

Two phase-locked pulses in direction q_1 produce interference effects depending on the delay τ_1. When the two levels are decoupled from the phonons ($g = 0$), the FWM signal in Eq. (6) oscillates as $\sin^2(\epsilon\tau_1/2)$, and is thus suppressed when the induced polarizations are exactly out of phase, i.e. $\epsilon\tau_1 = (2n + 1)\pi$.

In the strong coupling regime, the first two terms on the right-hand side of Eq. (6) correspond to the separate responses of each pulse which are peaked at $\tau = nT_{ph}$ and $nT_{ph} - \tau_1$, respectively. The third term describes the interference between polarizations and has its maximum at $\tau = nT_{ph} - \tau_1/2$ in between the two other peaks. Depending on its phase whose main contribution is $\epsilon\tau_1$, the interferences will be destructive or constructive.

Figure 2 shows the integrated FWM signal as τ_1 is tuned within a narrow range around $\tau_1 = 41\pi/\epsilon = 0.23T_{ph}$. At time $\tau_1 = 0.227T_{ph}$ the different contributions in Eq. (6) add up constructively to form a single broad peak at $\tau = T_{ph} - \tau_1/2$. At $\tau_1 = 0.231T_{ph}$, however, the destructive interference splits the peak into two bumps separated by a time $\tau \simeq 0.4T_{ph}$. The separation exceeds τ_1 because the interference suppresses the signal in the whole range of τ where the peaks $G(\tau, 0)$ and $G(\tau + \tau_1, 0)$ have a significant overlap.

The splitting of the phonon peaks was interpreted in Ref. 7 as a doubling of frequency due to two-phonon processes since the peaks were separated by approximately $T_{ph}/2$ for the values of τ_1 investigated. Within the two-level model, however, the additional peak that appears due to destructive interference, clearly moves with varying τ_1 and cannot be interpreted as a frequency doubling. Figure 3 shows the integrated FWM signal for various τ_1 tuned as to produce well separated doublet peaks, P^- and P^+, for time delays close to $\epsilon\tau_1 = 31, 41, 51$ and 61π. The P^- peak follows the dispersion $T_{ph} - \tau_1$ as illustrated by the inset which shows the positions of the maxima τ_{max} as a function of τ_1 for coupling strengths $g = 0.8$ and 1.5. While for $g = 0.8$ the rather broad peaks are splitted by more than τ_1, for $g = 1.5$ their positions approach the expected values of $\tau_{max} = T_{ph} - \tau_1$ and T_{ph} because the peaks have a reduced overlap.

The FWM signal for fixed τ but varying τ_1 shows bursts at multiples of the phonon period which are a clear signature of the strong coupling regime. Figure 4 shows the intensity of the first phonon peak $I(\tau = T_{ph}, \tau_1)$ rapidly oscillating with the interband frequency ϵ. The envelopes $I_\pm(\tau_1)$ of the oscillations shown as thick lines, vary exponentially with $x = \sin^2(\Omega\tau_1/2)$ for large g:

$$I_\pm(\tau_1) \propto 1 + \exp\left(-2\Gamma\tau_1 - 2g^2(1 + 4x - \sqrt{1+8x})\right) \pm$$
$$2\exp\left(-\Gamma\tau_1 - 2g^2(1 + 2x - (1 + 2x + \sqrt{1+8x})/2)^{1/2}\right) \quad (8)$$

For large g the envelopes exhibit bursts at multiples of the phonon period. The bursts are a non-perturbative effect involving all the many harmonics that contribute to the FWM signal. For an arbitrary time delay τ_1 the harmonics which are out of phase destroy the phase coherence induced by the pulses, and the oscillations are suppressed exponentially rapidly with τ_1. For resonant times τ_1 however, the different harmonics add up coherently and the phase sensitivity is recovered.

In conclusion we have studied the FWM response of a two-level system coupled to a single phonon mode focusing on the strong coupling regime. Analytical calculations of the integrated FWM signal provided new insight into the interplay between coherent control and dephasing through scattering processes: (i) The FWM signal has sharp peaks at multiples of the phonon period whose width decrease as $1/\sqrt{g}$ with increasing coupling. (ii) Two phase-locked pulses delayed by τ_1 give rise to two sets of peaks which add up coherently into a single set of bump at $nT_{ph} - \tau_1/2$ for $\epsilon\tau_1$ a multiple of π. When the two signals are out of phase, however, the peaks split into doublets at approximately $nT_{ph} - \tau_1$ and nT_{ph}. This picture contrasts with the interpretation of the coherent control experiments in terms of a frequency doubling. (iii) The FWM signal for fixed τ exhibits bursts of coherence at resonant values of $\tau_1 = nT_{ph}$, which are a clear signature of the non-perturbative regime and might be observed in experiments by measuring the intensity of the first phonon resonance as a function of τ_1.

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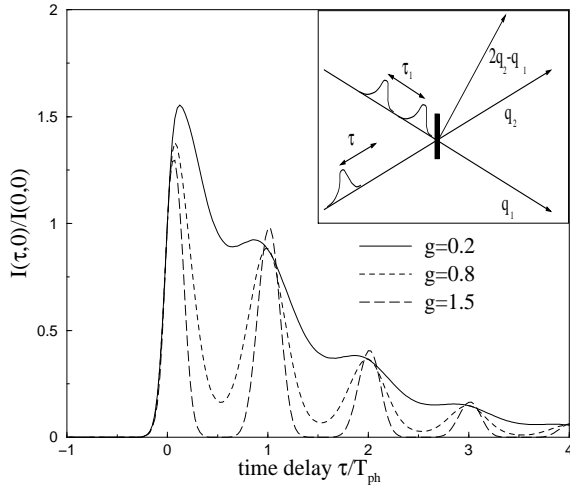[13] The corresponding nomenclature for time delays in Ref. 1 is $\tau = t_{21}$ and $\tau_1 = t_{11}$.

FIG. 1. Normalized integrated FWM signal $I(\tau, \tau_1 = 0)/I(0,0)$ as a function of delay τ in units of the phonon period T_{ph} for a single pulse in direction q_1, i.e. $\tau_1 = 0$. The weak modulation of the signal for $g = 0.2$ turn into distinct peaks at multiples of the phonon period with increasing coupling. The inset shows the setup for the coherent control where two phase-locked pulses delayed by τ_1 propagate in direction q_1. A third pulse in direction q_2 delayed by τ is diffracted along $2q_2 - q_1$.
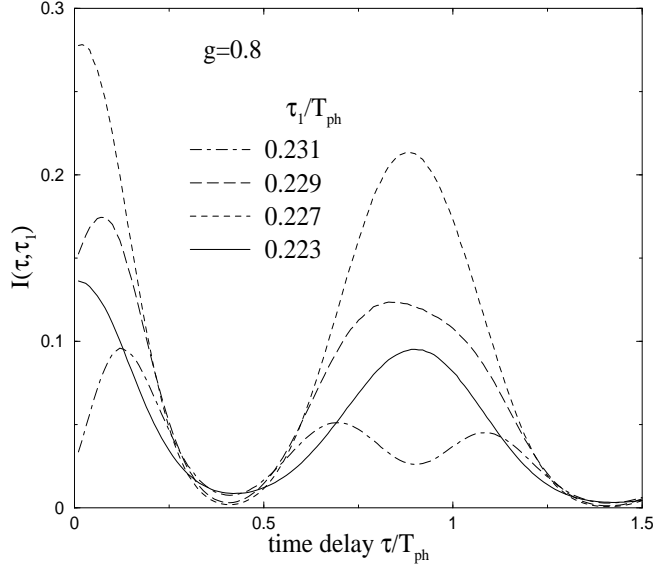

FIG. 2. Integrated FWM signal $I(\tau, \tau_1)$ as a function of delay τ for different τ_1 around $\epsilon\tau_1 \simeq 41\pi$. Constructive interferences between phase-locked pulses produce a single peak at $\tau = T_{ph} - \tau_1/2$ which splits into a doublet when τ_1 is tuned to achieve destructive interferences.
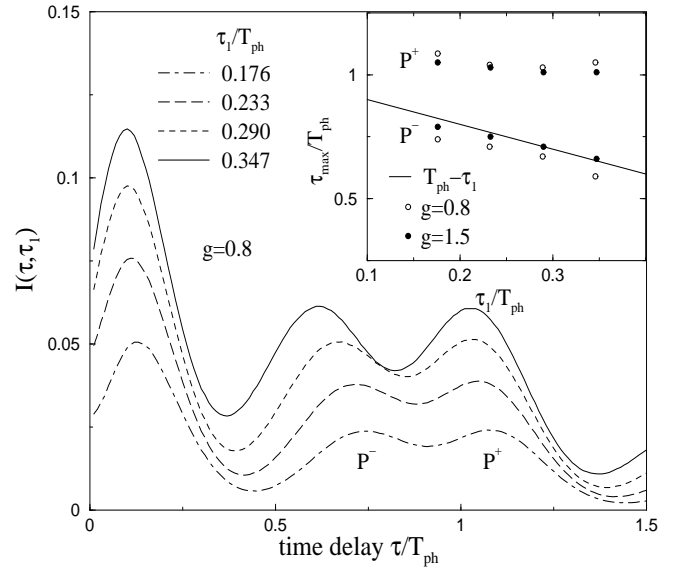

FIG. 3. Integrated FWM signal $I(\tau, \tau_1)$ for $g = 0.8$ as a function of time delay τ for different times τ_1. The inset shows the position of the maxima τ_{max} of the two peaks P^- and P^+ as a function of τ_1 for $g = 0.8$ and 1.5. With increasing coupling, the dispersion of the P^- peak approaches $T_{ph} - \tau_1$ plotted as the solid line.
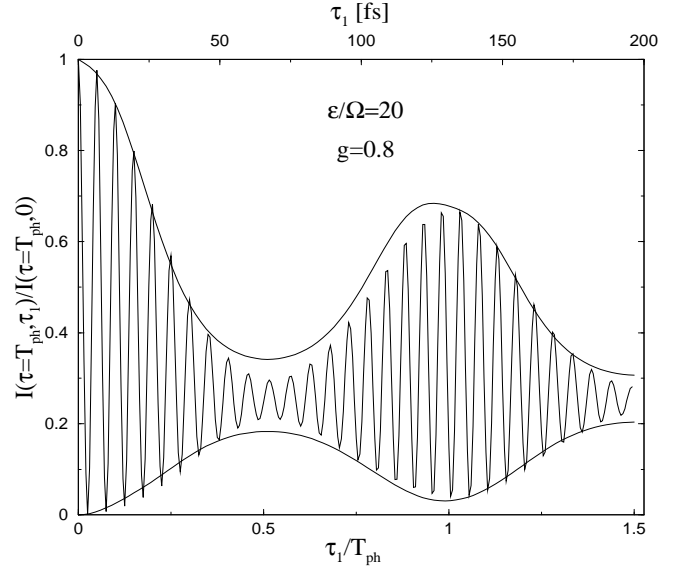

FIG. 4. Normalized intensity of the first phonon peak $I(\tau = T_{ph}, \tau_1)/I(\tau = T_{ph}, 0)$ as a function of delay time τ_1 between phase-locked pulses. The rapid oscillations of frequency ϵ which are first suppressed with increasing τ_1, show bursts at later times when τ_1 is a multiple of the phonon period T_{ph}. The envelopes of the oscillations are well described by Eq.(8) valid for large g (thick lines). For better display of the interference patterns, the frequency ϵ has been reduced by a factor of 4 from its value for ZnSe.